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X.

CONTRIBUTIONS FROM THE CHEMICAL LABORATORY OF HARVARD COLLEGE. — J. P. COOKE, *Director.*

THE RELATIVE VALUES OF THE ATOMIC WEIGHTS OF HYDROGEN AND OXYGEN.

By Josiah Parsons Cooke

and

Theodore William Richards.

Presented June 15, 1887.

Introduction.

Since the application by Dalton of the atomic theory to explain the definiteness of the combining proportions of the elementary substances of chemistry, these proportions have been generally regarded as the ratios of the weights of the atoms, and the values assigned to each element have been generally called atomic weights.

The conception was early suggested and advocated by Dr. Prout, an eminent physician of London during the first half of this century, that the elementary atoms were all aggregates of the atom of hydrogen, the lightest atom known. If this were true, it would of course follow that the atomic weights of the elements would all be multiples of the atomic weight of hydrogen; so that, if the weight of the atom of hydrogen were selected as the unit of the system, all other atomic weights must be multiples of this unit, and therefore whole numbers.

The facts known at the time (1815) were not inconsistent with this view; but as the methods of chemical analysis were improved, and the combining proportions determined with greater accuracy, marked discrepancies from Prout's hypothesis appeared. Still, so great was the hold which the conception had taken upon chemical students, that for a long time the nearest whole numbers to the combining proportions observed were accredited as the true value of the atomic weights, rather than the actual mean of the experimental results; and this practice is still followed in many standard publications, notably the "Jahresbericht über die Fortschritte der Chemie." In many cases the observed values were so near whole numbers that no important error in the calculation of analyses arose from this practice, the differences neglected being no greater than the uncertainties of analytical method, and this was especially true with regard to the larger atomic weights.

One exception to the theory, however, was so marked that it could not be overlooked, namely, the atomic weight of chlorine, which was capable of being determined with great accuracy; and all the determinations uniformly gave a result which was closely 35.5. This and a few similar cases suggested the idea, that, if the atomic weights were not even multiples of the received hydrogen atom, they might be multiples of the half or quarter hydrogen atom, which would simply amount to taking as the ultimate atom of material things a still smaller unit.

The well-known chemist, Dumas of Paris, was led by this view to undertake a redetermination of a large number of atomic weights, and many of the results then obtained are still accepted as authoritative.* As was to be expected, Dumas found a much closer agreement with this modified theory than with the original hypothesis of Prout; but obviously such evidence could have but little bearing on the general theory that the atoms were all aggregates of some common unit, for by taking that unit small enough, — even no smaller than the one hundredth of the received hydrogen atom, — all the atomic weights, even those most accurately determined, would be expressed by whole numbers within the limits of probable error.

Soon after, Stas of Brussels, a former assistant of Dumas, endeavored to set the question of Prout's theory at rest by an investigation which will be ever memorable for its extreme accuracy.† He selected for his investigation those elements whose combining proportions were capable of being determined with the greatest accuracy, and, working on large quantities of material, with every refinement which a full knowledge of analytical methods could suggest, he obtained results which it seemed impossible to reconcile with the theory in any way. This investigation, published in 1865, seemed at first to disprove the theory altogether.

Nevertheless, when Stas's results came to be collated, and as other determinations of similar accuracy came to be published, the fact appeared that a large number of the most accurately determined atomic weights stood to each other in the relation of whole numbers within the limits of accuracy of the most refined experimental work. The number of these cases was so large that it seemed highly improbable that the coincidences should be the result of chance.

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* Annales de Chimie et de Physique, 3d ser., lv. 129 (1859).

† Mémoires de l'Académie Royale de Belgique, xxxv. Also Ann. Ch. Pharm., Suppl., iv. 168.

This idea was prominently set forth by Professor Mallet of the University of Virginia, in his admirable paper on the Atomic Weight of Aluminum,* which was a striking illustration in point; and the same feature was also made prominent by Professor F. W. Clarke of Washington, after a careful review of all the determinations of atomic weights.†

The coincidences appear more striking if the values of the weights referred to are given in values of the oxygen atom assumed to be 16, as has been done by Professor George F. Becker in his digest of atomic weight determinations.‡ The following table from the writer's work on Chemical Philosophy will make clear the point in question.

ATOMIC WEIGHTS

MOST ACCURATELY DETERMINED.

Hydrogen	1.002	Chlorine	35.46
Lithium	7.01	Potassium	39.14
Carbon	12.00	Calcium	40.00
Nitrogen	14.04	Bromine	79.94
Oxygen	16.00	Silver	107.93
Aluminum	27.02	Antimony	119.92
Sodium	23.05	Iodine	126.85
Magnesium	24.00	Barium	137.14
Phosphorus	31 05	Thallium	204.11
Sulphur	32.07	Lead	206.91

This table includes all the atomic weights which up to 1882 could be regarded as known within one thousandth of their value, and with one or two notable exceptions there is no instance in which the value differs from a whole number by a quantity greater than the possible error, though not always the "probable error," of the processes employed in their determination.

Were these numbers wholly independent of each other and distributed by no law, we should expect to find every possible intermediate value, and the fact that they so nearly approach whole numbers cannot fail to produce on the mind the impression that there is some influence which tends to bring about this result.

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* Phil. Trans., 1880, p. 1003.

† Smithson. Misc. Coll.; Constants of Nature, Part V. p. 270.

‡ Smithson. Misc. Coll.; Constants of Nature, Part IV.

It may be that the discrepancies are due to unknown constant errors, which every experimentalist knows are greatly to be feared. Or it may be that there is in nature a tendency to whole multiples ; and this last view, if not compatible with our present conception of the atomic theory, may hereafter appear as one of the phases of a broader philosophy.

The force of evidence which such a distribution of values as the above table presents was brought home to the writer in his investigation of the atomic weight of antimony.* After eliminating various causes of error, he was enabled to determine with great accuracy the atomic weights of antimony, silver, and bromine, in one and the same series of experiments; and it appeared that this ratio was

$$120.00 : 108.00 : 80.00,$$

with a probable error of less than one in the last decimal place. Here then is a ratio of whole numbers within the one hundredth of a single unit. Since the ratio of the atomic weights of silver and oxygen have been determined with great accuracy, we can extend the above proportion to a fourth term, the atomic weight of oxygen, which appears also as a whole number, perhaps with a somewhat larger probable error. Still, we have not reached the unit of the system, and when we attempt to extend the ratio to the atomic weight of hydrogen, we find that the most probable value from all experiments hitherto made gives the ratio not of 16 to 1, but of 16 to 1.0025.

If now we wished to refer to the hydrogen unit the atomic weights of antimony, silver, bromine, and oxygen whose ratios of whole numbers had been determined as above, it was only necessary to divide all the terms of the above proportion by 1.0031, when we obtain the series of values given below the others, and all semblance to the hypothesis of Prout disappears, although of course the second series of numbers bear the same ratios to each other as the first: —

Antimony.	Silver.	Bromine.	Oxygen.	Hydrogen.
120.00	108.00	80.00	16.01	1.0031
119.60	107.66	79.75	15.96	1.00

The numbers in the lower of the two proportions appear as uncommensurable as Stas maintained that they were, and the same is true of most of the atomic weights, when given, as is usual in recent text-books, on the basis of the hydrogen unit.

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* Additional Experiments on the Atomic Weight of Antimony, Am. Acad. Proc., vol. xvii. p. 13, by Josiah Parsons Cooke.

When as the result of his investigation on the atomic weight of antimony there was presented to the writer the ratios of whole numbers as shown in the first of the above proportions, with the single exception of the atomic weight of hydrogen, the question was at once suggested: Is the ratio of the atomic weights of oxygen and hydrogen in fact that of 16 : 1.0025, as the general average of all trustworthy determinations hitherto made seems to indicate, or was there some constant error lurking in these results which caused the very slight variation from 16 to 1 required by the theory? In looking at the proportion thus displayed, it seemed as if the variation from the theory must be apparent, and he determined to ferret out the hidden error if possible. This investigation was undertaken in the autumn of 1883, but owing to the condition of the writer's sight the work has been greatly delayed.

No one can study the record of the investigations by which the ratio of the weights of the oxygen and hydrogen atoms have been determined, without receiving the impression that they are by no means decisive in regard to the theory we are discussing, and it is also equally evident that this ratio, if it could be fixed beyond doubt, would be a crucial test of the theory.

Previous Work.

The methods by which the atomic weights of oxygen and hydrogen have been determined may be divided into two classes; first, the direct method of determining the ratio in which the proportions of oxygen and hydrogen uniting to form water were actually weighed; secondly, the confirmatory method, to whose results small weight could be given independent of the first.

Among confirmatory methods we must unquestionably class the classical determinations by Regnault of the density of oxygen and hydrogen gases under normal conditions at Paris; that is, in so far as these determinations bear on the question of the ratio of the atomic weights.

According to the molecular theory the ratio of the densities of oxygen and hydrogen gases could only be the ratio of their molecular or of their atomic weights when both materials were in the condition of perfect gases, of which condition the test would be an exact conformity to Mariotte's law. Now, as Regnault himself elegantly demonstrated, oxygen and hydrogen gases at the standard conditions of temperature and pressure not only do not exactly obey Mariotte's law, but the deviations from the law in these two cases are in the opposite directions, oxygen gas being condensed by increasing pressure more, and hydrogen gas less, than the law requires. Hence theory would not lead us to expect that the ratio of the densities of these gases at the standard conditions would be the exact ratio of their atomic weights. But obviously it may be that this inference from the molecular theory is not legitimate, or it may be that the effect of the imperfect aeriform condition would not perceptibly influence the apparent atomic ratio; and hence the confirmation afforded by Regnault's results is of value.

In the same category we must class also the determination of the atomic weight of oxygen made by Thomsen of Copenhagen, who weighed the amount of water obtained by burning a measured volume of hydrogen gas. Here the reduction of the volume to weight involved a knowledge of values and conditions which could not be known with the greatest accuracy, and unfortunately the details of the experiments have not been published.

Again, we should class simply as confirmatory results deduced indirectly, and involving the values of other atomic weights, however accurately these subsidiary values may be supposed to be known; such, for example, as Stas's determination of the amount of chlorine in ammonic chloride.

Turning now to the actual direct determinations of the combining proportions of oxygen and hydrogen, there are only two which are of any present value. Of these by far the most important is the classical investigation of Dumas, "Researches on the Composition of Water." * This is one of the most memorable investigations in the history of chemistry, and its general principles are known to every student of the science. An indefinite amount of hydrogen was burnt by means of cupric oxide; the amount of oxygen consumed was determined by the loss of weight of the combustion tube, and the amount of water formed was collected and weighed directly. The experiments were on a very large scale, the amount of water produced varying from 15 to 70 grams. The greatest care was taken to insure the purity of the materials used, every known experimental means was employed to secure accuracy, and all necessary corrections were applied to the results. Estimated on the system at present in use, the value of the atomic weight of oxygen obtained by Dumas as the mean of nineteen

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* Ann. de Chim. et de Physique, 3d ser., viii. 189 (1842).

determinations was 15.9607, with a probable error of ±0.0070, the highest value being 16.024, and the lowest 15.892.

The investigation of Erdmann and Marchand* was far less extended, and some of the precautions taken by Dumas were neglected because deemed unnecessary. No pains seem to have been taken to obtain pure cupric oxide, and the material used in several of the determinations was described as "kaufliche Kupferglühspan," while that used in the others was obtained by igniting cupric nitrate; and no proof is adduced in either case of the purity of the material employed.

The results are divided into two groups, and in the experiments of the second group the air was exhausted from the combustion tube before weighing; but it appears from the paper that the marked difference between the two series of experiments depended rather on the character of the cupric oxide, and on varying conditions used, than on this circumstance. The first series of four results, when averaged, gave the value 15.937, with a probable error of ±0.0138, while the mean of the second series was 16.009, with a probable error of ±0.0030. The study of the paper, however, does not confirm the expectation that the results of the second series are more trustworthy; for the closer agreement and smaller probable error appear to be the result of the identity of conditions, which was maintained in this series, but not in the first. Judging from the paper, we should be inclined to place most reliance on the first series, in which the conditions of the experiments were varied, rather than on the second, which seems obviously to be influenced by some constant error.

The results, then, thus far obtained, are as follows: —

Direct Determinations.

Dumas (nineteen determinations)	15.9607 ± 0.0070
Erdmann and Marchand (first four)	15.9369 ± 0.0138
" " " (second four)	16.0095 ± 0.0030

Confirmatory Determinations.

Dumas and Boussingalt † (gas densities)	15.954 ± 0.031
Regnault ‡ (gas densities)	15.961 ± 0.0044
Thomsen § (not fully described)	15.960

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* Journ. f. Prakt. Chem., 1842, vol. xxvi. p. 461.

† Compt. Rend., xii. 1005; also Constants of Nature, Part V. p. 6.

‡ Compt. Rend., xx. 975; also Constants of Nature, Part V. p. 6.

§ Berichte der deutsch. Chem. Gesell., 1870, p. 928; also Constants of Nature, Part V. p. 8.

The one process by which the relative combining proportions of oxygen and hydrogen have been hitherto directly determined is open to serious criticism. In the first place, the circumstance that the weight of the hydrogen is eight times smaller than that of the oxygen, and that this weight has only been estimated by difference, is exceedingly unfavorable to the accuracy of the process. It can easily be seen, that, in order to establish a ratio like 1 to 8, the highest accuracy demands that each term of the proportion should be known to an equal degree of exactness. Thus if in a given experiment we have 8 grams of oxygen uniting with 1 gram of hydrogen, it is of no avail to weigh the oxygen to the tenth of a milligram, unless we can weigh the hydrogen to the same proportionate degree of accuracy. For an error of eight tenths of a milligram in the weight of the oxygen, or an error of nine tenths of a milligram in the weight of the water, will have no more influence on the ratio we are seeking than an error of one tenth of a milligram in the weight of the hydrogen. Now, in the process we are discussing the weight of the water can be determined to within a few tenths of a milligram; that is, with all the accuracy with which our problem requires that the larger term of the proportion 8 to 1 should be known. It is quite different with the weight of the oxygen. This last is found by weighing the glass combustion tube containing cupric oxide before and after the experiment, and between the two weighings the tube is heated to a low red heat for several hours while a stream of hydrogen gas is passing through it; and there are several causes which may lead to the variation of these weights, independent of the oxygen which has been used up in the process. We shall allude to some of these causes below, but their effect would be comparatively unimportant if they only led to a small error in the observed weight of the oxygen. Unfortunately their effect is not thus limited; for when, in order to find the weight of the hydrogen, we subtract from the weight of the water, which may be regarded relatively as accurately known, the weight of the oxygen, which may be for the causes referred to slightly erroneous, the whole error appears in the weight of the hydrogen thus found, and in the opposite direction. If, for example, the weight of the oxygen is too large, the weight of the hydrogen will be too small by exactly the same amount; and although the error may be an inconsiderable part of the weight of the oxygen, it may be a very appreciable quantity in the weight of the hydrogen.

On the other hand, if a means could be devised for weighing the hydrogen, leaving the oxygen to be determined by subtracting this smaller weight from the weight of the water, then a small error in the observed weight of the hydrogen would have no appreciable effect on the weight of the oxygen.

Dumas fully recognized the source of error to which we have referred, and in his paper on the subject wrote what may be translated as follows: —

"Of all analyses presented to a chemist, that of water is the one which offers the greatest uncertainty. Indeed, one part of hydrogen unites with eight parts of oxygen to form water, and nothing would be more exact than the analysis of water, if we could weigh the hydrogen as well as the water which results from its combustion. But the experiment is not possible under this form. We are obliged to weigh the water formed, and the oxygen which was consumed in producing it, and to determine by difference the weight of the hydrogen which has entered into combination. Thus an error of $\frac{1}{900}$ in the weight of the water, or of $\frac{1}{800}$ in the weight of the oxygen, is equivalent to an arror of $\frac{1}{80}$ or $\frac{1}{90}$ in the weight of the hydrogen. Let these two errors be in the same direction, and the total error will amount to $\frac{1}{40}$."

In the second place, however carefully the exterior surface of the combustion tube may be guarded, it is impossible that the contents of the tube should bear the same relations to the surrounding atmosphere before and after the combustion. We begin with a tube containing cupric oxide in different states, and we end with it containing reduced copper, whose condition will vary more or less with the character of the oxide employed; and the power of these materials to occlude air or hydrogen is an unknown quantity in our experiment. That it is an appreciable quantity is evident from several incidental observations.

Dumas endeavored to avoid any source of error arising from this cause by exhausting the combustion tube before weighing it, but he himself expresses a doubt whether a trace of hydrogen might not have been left. Erdmann and Marchand in part of their experiments resorted to the same expedient, but their results obviously vary with the condition of the cupric oxide employed; and the following remarks of Schutzenberger, in a discussion of the variability of the law of definite proportions before the Chemical Society of Paris in 1883, as quoted in the "American Journal of Science," 3d series, Vol. XXVI. page 65, have an important bearing on the same point: —

"When water is synthesized by reduction of a known weight of CuO, by weighing the reduced Cu and the water formed, it is found that the ratio of O to H is not constant, but varies with the state of division and of saturation of the oxide, the duration of contact of the water formed with the oxide, and with the temperature, from 7.95 to 8.15. The latter value is obtained with a saturated and divided oxide filling the tube, the former with oxide in lumps filling the tube for a space of 25 cm. With a larger empty space the ratio has fallen to 7.90. When the synthesis of water is effected by weighing the hydrogen consumed (as by dissolving a known weight of zinc in HCl) and the water formed, the ratio differs according to the contents of the combustion tubes. If it contains granular CuO of a length of 80 cm. heated to redness, the ratio O : H is 7.96 to 7.98 to 1; at a low temperature, 7.90 to 7.93; if the CuO is replaced by $PbCrO_4$, from 7.89 to 7.93."

In addition to all this, impurities in the oxide of copper might have a serious influence on the result. As before said, Erdmann and Marchand speak of using "kaufliche Kupferglühspan"; but in our work we could find no commercial cupric oxide which did not contain a marked amount of arsenic. We examined a number of specimens coming from the best German and American dealers, and there was not a single instance in which we did not find arsenic, and even when the material was marked "purissimum." In some cases the amount of arsenic was so great, that, after successive reductions and oxidations, abundant crystals of arsenious oxide collected at the exit end of the combustion tube. It is unnecessary to add that the hydrogen used was free from all such impurity.

For our own experiments, of which the results are given below, not only was the oxide of copper prepared from absolutely pure electrolytic copper, but also, as will be shown, the combustion tube was left at the end of the determination as it was at first, and the same tube was used for a number of experiments.

Apparatus for weighing Hydrogen.

In entering on a new investigation of the oxygen and hydrogen ratio, it was evident at the outset that no advantage was to be gained by multiplying determinations by the old method. The only hope of improvement lay in finding some method of weighing the hydrogen with sufficient accuracy; and it was essential to determine this weight to within one ten-thousandth, or at least one five-thousandth, of its value.

A gas can only be weighed by enclosing it in a glass globe, or some similar receiver, and hydrogen is so exceedingly light that its total weight can only be a very small fraction of the weight of the containing vessel. Moreover, as the buoyancy of the air is fourteen and one half times as great as the weight of the hydrogen, the variations in buoyancy caused by changing atmospheric conditions have an all-important effect on the apparent weight. The late Professor Regnault, of Paris, devised, however, a very ingenious method of compensation. which could readily be applied in this case. It consisted in balancing the globe containing hydrogen, hung to one arm of the balance, by a second globe of exactly the same external volume and made of the same material, hung to the opposite arm; and so arranging the balance case that they should hang in the same enclosure, and therefore be equally affected by atmospheric changes. This method was applied in the problem before us; and after a number of trials it was found possible to make the compensation so accurate that under good conditions the weight of a globe holding five litres of gas did not vary more than one tenth of a milligram through large changes of temperature and pressure. In order now to weigh hydrogen with this apparatus, it was only necessary to exhaust the air from the glass receiver, and, after balancing it as described, to fill it with pure gas, when the increased weight — less than half a gram with our apparatus — was the weight of hydrogen required.

The balance employed was an excellent one, made about twenty years ago by Becker and Sons, of New York. With a load of five hundred grams in each pan, it turns very perceptibly with one tenth of a milligram, and shows this small difference of weight with very great constancy.

The globe and its counterpoise were hung from hooks soldered to the bottoms of the pans by means of wires which swung freely through small holes made for the purpose through the bottom of the balance case, and also through the top of the shelf on which the case stood. The enclosure in which the globe and its counterpoise hung was a box made of tinned iron fastened to the bottom of the shelf, and having doors in front like an oven, through which the globe could be removed or hung in position. This case was coated with lampblack on the inside, in order to secure uniformity of temperature; and the air was kept dry by means of two large dishes of sulphuric acid, placed on shelves at the top of the case. We first placed the sulphuric acid dishes on the bottom of the case in the usual way, but we found it impossible thus to secure a uniform condition of the atmosphere within; and as moist air is necessarily lighter than an equal volume of dry air at the same temperature and pressure, it is obvious that any drying material will have the greatest efficiency when placed near the top of the space to be protected. The tin box was itself enclosed in a cupboard, but not otherwise protected; and the balance case was surrounded by curtains, in order to shield the beam from radiation.

With the apparatus so arranged, it was found possible to obtain most satisfactory and concordant results to tenths of a milligram when the change in the temperature of the balance-room was not very rapid; but any sudden changes produced by artificial heating would cause slight currents of air in the interior of the case, whose effect became very sensible, but whose influence we were able to eliminate. The best series of results, however, (the second series in the table below,) was obtained during the month of June, when there was no artificial heat in the building, and the temperature varied but little during day and night.

The globe used for holding the hydrogen — shown in Fig. 1 in about one sixth of its actual dimensions — has an interior capacity of 4961.5 cubic centimeters, and weighs 570.5 grams. The cap with the connecting tubes was ground into the neck, and this joint, as also the stopcocks, was so carefully made that there was absolutely no leakage, and the globe would hold a vacuum indefinitely; as was shown frequently by its remaining hung on the balance for weeks together when exhausted without change of weight. The apparatus was made by Emil Greiner, of 79 Nassau Street, New York, whose careful workmanship greatly contributed to the success of our investigation. The details of the stopcock are shown at the sides; and it will be noticed that, besides the direct way, there is a side way through the plug of the stopcock independent of the first, by which, when the stopcock is closed, a connection is established with the base of the cock, through which the gas may escape.

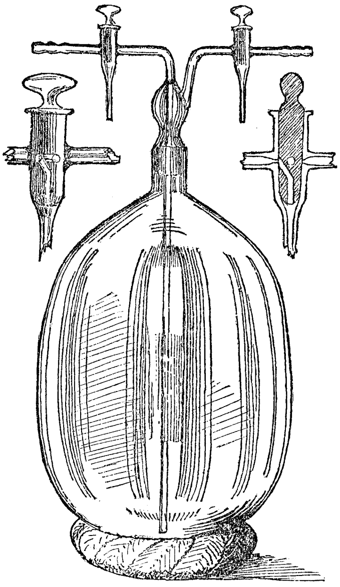

Fig. 1.

Assume now that the interior of the globe has been exhausted, and a gas current established through this side aperture from one of the generators employed. On turning the stopcock, the side aperture is first closed, and then the direct way slightly opened, so that all the gas evolved now passes into the globe; and it was found possible to regulate the current with such nicety as not to cause any sudden changes of tension in the generator, which was always provided with an overflow by means of which the tension could be watched, and according to which the stopcock was regulated.

The filling of the globe was one of the critical points of the determination. It generally occupied from one to two hours, and during all this time it was necessary, with the hand on the stopcock, carefully to watch the tension at the overflow already mentioned, and represented in Fig. 5, and Figs. 7 and 8 of Plate. From the beginning to the end of the operation there was a greater tension in the generator than in the outside air, by about one inch of mercury. When the connection was once established between the generator and the globe, there was absolutely no leakage through the side way, as was tested in several cases by dipping the mouth of the tube at the base of the cock under mercury.

The whole process of weighing the hydrogen was finally reduced to the following manipulation. The globe was connected by means of a rubber hose with a rotatory air-pump having automatic valves, made by E. S. Ritchie of Boston. It was then exhausted to within 1 mm. of mercury. Next, closing the cock and disconnecting the globe, it was cleaned with distilled water and fine cotton cloth; at least, this was done five or six times during the determinations. But as the globe when out of the balance case was always protected by a cylindrical tin box with a cover, from which the exit tubes projected, it was usually only necessary to clean the exit tubes in this careful manner, simply dusting off the globe with a large camel's-hair brush, before hanging it in the balance case. In this part of the operation it was necessary to take care not to communicate to the globe a charge of electricity by rubbing it with a perfectly dry cloth.

The globe was hung on a wire stirrup, which caught the exit tubes, as the glass joint was sufficiently strong to support the weight of the globe. The globe was so nearly balanced by its equipoise that when exhausted it only required about one decigram to establish equilibrium. The time required to attain perfect equilibrium varied with the conditions. If the glass had been previously cleaned as described above, perfect equilibrium might not be reached for forty-eight hours, or even longer, while if the glass had only been dusted, twelve hours were generally sufficient.

After the tare had thus been taken, the globe was removed from the balance, placed in the protecting case, and filled with hydrogen as just described. The inlet tube, to which a rubber connector had been attached, was scrupulously cleaned as before, and the globe was again dusted and hung on the balance. During all these transfers the globe was always handled with clean cotton cloth, and the hands never came in contact with the glass. The increased weight was now the weight of the hydrogen; and as the volumes equipoised were exactly the same, and the additional weight was represented by less than five tenths of a gram of platinum, any correction for the buoyancy of the atmosphere is unessential.

Combustion Apparatus.

The apparatus by which the combustion of the hydrogen was made is represented in the Plate accompanying the paper (Fig. 6). It is made up of a series of small combustion furnaces, which are a modification of a kerosene-oil stove called " the American," very much used in the United States. This stove, as adapted to chemical uses by the writer, is shown in Figs. 2 and 3, and it has proved of great value, not only for elementary chemical experiments in school courses, where illuminating gas is not to be had, but also in a well equipped chemical laboratory. The stove is made for burning kerosene oil, but alcohol can also be burnt in it with decided advantage for chemical work. The figure of the stove has been drawn to about one sixth of the actual size.

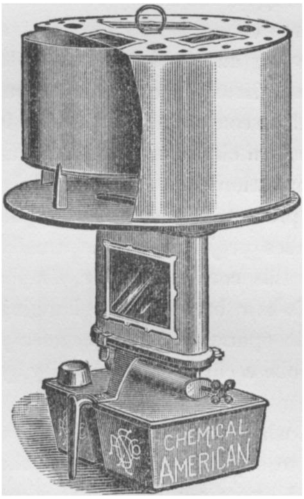

Fig. 2.

In the figure of the combustion apparatus (Fig. 6, Plate) it will be noticed that the globe, protected by its case, stands about in the middle of the line. By means of a suction pump attached to the extreme right of the apparatus, a current of air is maintained through the whole length. Beginning now at the extreme left, the air first passes over reduced copper, and is deprived of its oxygen. It next passes over cupric oxide, by which any traces of hydrogen that had remained occluded by the reduced copper, or any traces of hydrocarbons in the air itself, are burnt. It next passes through caustic potash bulbs, and then through a system of driers, meeting successively calcic chloride, sulphuric acid, and phosphoric pentoxide. It now enters the globe through the inlet tube reaching to the bottom, carrying before it the hydrogen into the combustion furnace.

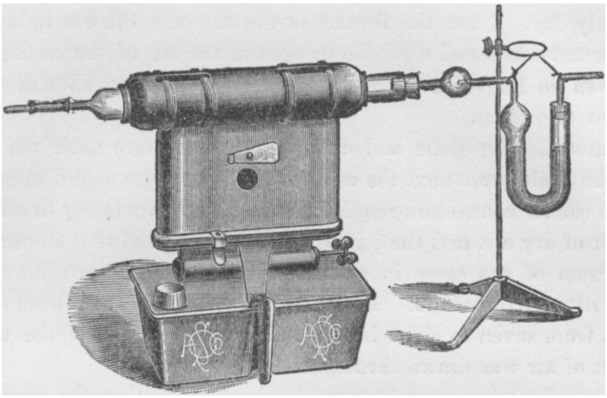
Fig. 3.

The water from the combustion was collected in a condenser, — the details of whose construction are represented in Fig. 4, — which was shielded from the furnace by a screen of asbestos paper. Nine tenths of the water was condensed in the middle tube, and all but the last traces of the aqueous vapor were absorbed by the sulphuric acid through which the air subsequently bubbled up at the bend of the U tube, between glass beads, which broke the ascent and divided the bubbles. With this condenser was connected a U tube containing phosphoric pentoxide, which absorbed the last traces of the aqueous vapor, seldom, however, gaining in weight more than two milligrams during a combustion lasting from seven to eight hours. Then follows a safety tube containing calcic chloride (or in some cases phosphoric pentoxide), to prevent any reflex diffusion, and finally an adaptation of the principle of Mariotte's flask to regulate the velocity of the air current. It will be seen that the open mouth of the central tube of this last apparatus dips under the mercury in the tall jar, so that by raising or lowering it the strength of the current could be exactly regulated.

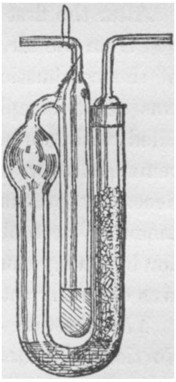
Fig. 4.

Before beginning a combustion, the place of the hydrogen globe was supplied by a straight piece of glass tubing, and the air current maintained through the heated combustion tube until the cupric oxide was perfectly dry. Next the furnace at the extreme left was lighted, and the current continued until the oxygen in the air of the drying tubes had been so far replaced by nitrogen as to remove all risk of subsequent explosion.

Meanwhile the globe and condensing tubes were made ready, and first the globe and next the condensing tubes were placed in position, all the rubber connectors required having been previously dried in the current of dry air, and the joints were so contrived that subsequently the stream of gas came in contact with the smallest possible surface of the rubber connectors. The combustion lasted, as has been already stated, from seven to eight hours, and during this time quite a rapid current of air was drawn through the apparatus.

Our preliminary experiments plainly showed that the rapidity of the current within practicable limits had no appreciable effect on our results, and this is due to the fact that the current entered the globe and left the condensers under precisely the same conditions. More than nine tenths of the water was condensed during the first half-hour, the drops falling regularly from the mouth of the inlet tube, and after two hours all traces of cloudiness disappeared from this tube or its connections, showing that the air coming over was perfectly dry. The water thus collected was absolutely clear and limpid.

After the first hour the combustion furnace used for removing oxygen from the air, at the extreme left, was taken away, and by the end of the combustion the reduced copper in the combustion tube proper was again completely oxidized, leaving the globe and all the tubes filled with normal air, as at the beginning of the process. It only remained now to remove the condensers, and reweigh them with all necessary precautions. At both weighings the barometer and thermometer were observed, and the small, usually insignificant correction for buoyancy caused by a change in the atmosphere during the interval was carefully estimated.

The question of the time of running the combustion after the production of water had sensibly ceased was one that was carefully considered. The time mentioned above — eight hours — was far outside the necessary limits, and was reached only after a large number of experiments. That a very long continuance of the current after the combustion was practically ended was unnecessary, was clearly shown by several circumstances. In the first place, the duration of the combustion beyond the limit we have named made no difference in our results, as was repeatedly shown. Again, in one instance, after detaching the condensation tubes and weighing them, they were again put in place and the combustion continued three hours longer, during which time the tubes gained no appreciable weight. In another instance, when a suspicion arose that possibly some hydrogen might be occluded on the walls of the globe, the condensation tubes having been dismounted and weighed as before, the globe was also dismounted and heated over the free flame of a Bunsen lamp to as high a temperature as the glass would safely bear, over 300° C., and then, the apparatus having been remounted, the combustion was continued for one hour. Here again the condensation tubes gained only a small fraction of a milligram in weight, an effect which might easily be accidental, and which was wholly without influence on the result.

Apparatus for preparing Hydrogen.

In the preliminary determinations, the hydrogen was drawn from a large copper generator charged with zinc and dilute sulphuric acid. The zinc and sulphuric acid were wholly free from arsenic, and of the best quality, but not absolutely pure; and the writer depended upon an elaborate system of washers and driers for purifying and drying the gas. He found the greatest difficulty in removing the last traces of sulphurous oxide, which hydrogen prepared in this way always carries. The presence of this trace cannot be detected by litmus paper, but is immediately indicated by the production of hydric sulphide when the gas is passed over heated platinum sponge; and by interposing a tube containing platinum sponge maintained at a low red heat, followed by a set of potash bulbs, this impurity can be entirely removed. It can also be removed by washing with a strong solution of potassic hydrate alone, if the gas remains long enough in contact with the solution. It was found, however, that a series of potash bulbs was insufficient for this purpose; but two of the long washers represented in the background of Fig. 5 and in Figs. 7 and 8 (Plate), where the gas in small bubbles travels up a tube 5½ feet long, are sufficient to remove the sulphurous oxide from even a quite rapid current of hydrogen gas. A series of preliminary determinations was made with hydrogen gas thus prepared and purified, and it was obvious from an inspection of the results, as well as from the difficulties which were experienced in keeping all the joints of this complicated apparatus tight, that the irregularities arose from the diffusion of the air into the hydrogen at some one or other of these joints. It was therefore next sought to simplify the apparatus, and to depend upon the purity of materials rather than on the completeness of purifying methods for obtaining pure hydrogen. Meanwhile, for reasons stated below, the writer had reduced very materially the scale of his operations, and this rendered unnecessary the large generator we had first employed.

The second apparatus that was constructed is represented in Fig. 5. Of this, the generator, in which hydrogen is made from pure zinc and hydrochloric acid, is the same as that described by Julius Thomsen.* The Wolff bottle is filled with pure granulated zinc, and the upper bottle contains pure hydrochloric acid diluted about one half. By means of a glass stopcock the acid is allowed to flow into the zinc drop by drop, and in this way the current of hydrogen can be quite closely regulated. Tubes protected by stopcocks are provided for adding fresh charges of acid, and for drawing off the solution of zinc chloride; also a tube connecting the upper part of the two bottles enables the operator to effect these transfers without introducing any air. The gas from this generator passed first through a long potash tube inclined at about 10° to the horizon, then through a tube about three feet long filled with calcic chloride, then through a glass tower filled with glass beads drenched with sulphuric acid, and lastly through a second tower filled with phosphoric pentoxide. As many of the joints as possible were made by fusing together the glass, and all the others were protected by a cement consisting of equal parts of pitch and gutta-percha. It will be noticed that an overflow is provided at the point where the potash tube connects with the calcic chloride tube,

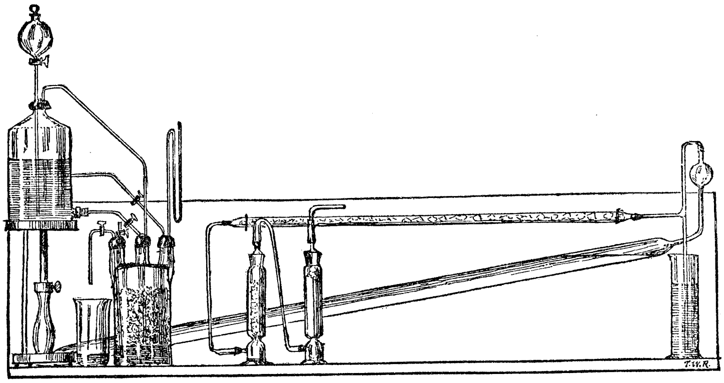

Fig. 5.

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* Thermochem. Untersuch., vol. i. p. 28.

the open mouth of the overflow tube dipping about six or eight inches deep under concentrated sulphuric acid. This overflow indicated the least change of tension of the hydrogen in the apparatus, and would have shown the least leak if it had existed; but the apparatus as thus constructed remained absolutely tight so long as it was in use.

With the hydrogen drawn from this apparatus, the first determinations were not wholly satisfactory, and the cause of error was traced to the air dissolved in the dilute hydrochloric acid with which the generator was charged. In all the succeeding determinations the greatest pains was taken to remove the last traces of air by boiling the dilute acid, and allowing it to cool in a stream of hydrogen; and as additional precaution, while the solution was still warm, the gas was exhausted from the containing vessel and pure hydrogen run in, several times in succession, the pure acid being finally conveyed into the generator entirely out of contact with the air. The need of all these precautions will be seen when it is considered how small an admixture of air or of nitrogen will materially influence the weight of the hydrogen. If only one ten-thousandth of the volume of the hydrogen were replaced by air during the process of filling the globe, this would cause an apparent increase of weight in the hydrogen of five tenths of a milligram, and that, other things being equal, would reduce the atomic weight of oxygen two hundredths of a unit.

The precautions used in filling the globe have already been described in detail, and with hydrogen from the apparatus, constructed and charged as just described, were made the five consecutive determinations whose results are given as of the first series in the table on page 173. These, and all the determinations given in the table, were made by the writer's pupil and assistant, Mr. Theodore William Richards, to whose experimental skill the success of the investigation is largely due, and without whose assistance the work could not have been completed in the present condition of the writer's sight. The mean of these first five results, as will be noticed, is but little less than that obtained by Dumas, and the probable error, ±0.0048, is considerably less than that of Dumas. In order to understand how this result appeared to the writer, it must be remembered that he started with a certain prepossession in favor of the hypothesis of Prout, based on his previous work on antimony; and, furthermore, that the effect of the causes of error which had been encountered and overcome all tended to lower the atomic weight; and the result obtained was a maximum which had been reached after every known precaution had been taken. But although this maximum was essentially the same as that obtained by Dumas by an obviously less direct and less accurate method, yet it was still possible that there might be some constant error, and that some cause might yet be found which would raise the maximum by the forty-six thousandths required to give the whole number 16. It was true that the probable error was only about one tenth of this difference; still, as the materials had been purified, the maximum had constantly risen, and the theoretical limit was in sight. In reviewing the work, it was obvious that the degree of accuracy of the methods used for determining the weights both of the hydrogen and of the water was so great, that no possible error in these values could account for the difference in question. This would imply an error of 1.2 milligrams in the weight of the hydrogen, and of 10.8 milligrams in the weight of the water, and the possible error of a single determination — leaving out of account the reduced probable error of the average value — was far within these limits. If there was a constant error, it must result from the want of purity of the hydrogen gas, and we therefore determined to try another method for preparing the hydrogen.

The apparatus next used is represented in Fig. 7 (Plate), and differs from the last only in the generator. Here the generator is a three-necked bottle having a capacity of about two litres, filled to about one eighth of its capacity with a semifluid amalgam of mercury and pure zinc. On this rests dilute hydrochloric acid, containing about twenty per cent of HCl, nearly filling the bottle. Into this acid dips a platinum electrode, while a straight glass tube passing through the middle neck and dipping under the amalgam gives the means of establishing an electrical connection between the large platinum plate which forms the negative electrode and the amalgam. In addition, a siphon tube for drawing off the acid when saturated with zinc, a funnel tube for introducing a fresh charge, and an exit tube, all well cemented to the several necks of the bottle, complete the generator. When the electrical connection is broken, all chemical action ceases, but on connecting by a wire the platinum electrode with the amalgam, a very steady but slow evolution of hydrogen gas takes place, which can be regulated with the greatest nicety by varying the resistance of the connecting wire. On interposing two cells of a Bunsen battery the evolution of gas becomes very rapid. Besides its special use in this connection, the apparatus will be found of great value as giving an absolutely constant source of pure hydrogen whenever required. In charging the generator with acid, the same care was taken to exclude every trace of air as with the previous apparatus, and with hydrogen thus prepared a second series of five consecutive determinations was made, whose results are given in the table on page 173, below those of the first series; and it will be noticed that, while the average value obtained is essentially identical with the previous result, the agreement of the several determinations is more close, and in consequence the probable error is reduced more than one half. A closer agreement under the circumstances could not possibly be expected.

Such a striking confirmation of the previous result seemed very conclusive, and the very small probable error indicated a command of the method which was very satisfactory. Still, it could not be proved that there might not be a constant impurity in the hydrogen used. As the hydrogen had passed every possible chemical test unimpeached, the only possible impurity that could be suspected was nitrogen, and Mr. Richards therefore made a careful spectroscopic examination, searching for the more conspicuous nitrogen lines in the spectrum obtained by passing an induction current through a rarefied atmosphere of the gas from the generator just described; but not the faintest trace of any of these lines could be seen.

Still, as in the electrolytic method of preparing the hydrogen the same materials, hydrochloric acid and zinc, were used as in the first series of experiments, it was determined to procure hydrogen by a wholly different chemical process, using the well-known reaction of metallic aluminum upon a solution of potassic hydrate.

The purest aluminum sheet that could be obtained in the American market was procured for the purpose, and the apparatus represented in Fig. 8 (Plate) was used for generating the gas. The generator here was a simple flask holding a strong solution of chemically pure potassic hydrate, and the aluminum was introduced in small pieces through a large open tube, — dipping under the surface of the solution, — the liquid being maintained at a level near the open mouth of the tube by the tension in the interior of the apparatus. The small strips of aluminum were carefully cleaned, and caused slowly to slide down the tube; the evolution of hydrogen from the surface began as soon as the strips of metal touched the liquid, and became very active in the tube before they dropped into the flask. And this action insured the removal of any traces of air which might adhere to the surface. In this apparatus the long caustic potash washer was not used, as being no longer necessary, and the gas was passed through caustic potash bulbs to remove the spray, and then through a calcic chloride tube, and over sulphuric acid and phosphoric pentoxide, as before.

With hydrogen thus prepared, the six determinations of the third series in the table were made; and it will be seen that the average of the results is a value which is essentially identical with the average values from the other two series. The probable error in this last series is larger than in the second, although still very small; but the difference is due, as the note-books plainly show, to the different conditions under which the two series were made. As before stated, the compensation of the balance was perfect, and the apparent weight of the globe did not alter by a tenth of a milligram, even with wide variations of temperature and pressure, so soon as those changes became constant. But when the changes of temperature in the balance-room were rapid, currents of air were established in the case, however great care was taken in protecting it, which rendered the apparent weight irregular to the extent of one or two tenths of a milligram; and the third series was made under less favorable conditions in this respect than the second. This point is illustrated by the following notes of two determinations, which are given in full, in order that all the circumstances connected with the determinations may be seen.

SERIES II. *Determination* 5.

Weighings of the globe: —

					Grams.
Exhausted.	June 6th,	6.00 P. M.	Tare	=	0.1960
	" 7th,	7.25 A. M.	"	=	0.2011
	" "	8.30 A. M.	"	=	0.2011
	" "	11.20 A. M.	"	=	0.2011
	" "	2.00 P. M.	"	=	0.2011
					0.2011
Filled with Hydrogen.	June 7th,	7.20 P. M.	Tare	=	0.6100
	" 8th,	8.00 A. M.	"	=	0.6156
	" "	10.00 A. M.	"	=	0.6156
	" "	12.15 P. M.	"	=	0.6154
	" "	7.50 A. M.	"	=	0.6155
	" "	9.30 A. M.	"	=	0.6155
	" "	11.40 A. M.	"	=	0.6155
					0.6155

Weight of Hydrogen taken = 0.6155 — 0.2011 = 0.4144 gram.

The combustion was started at 11 A. M., and stopped at 6 P. M.

Weight of P_2O_5 tube,

before combustion	= 48.2499	$h = 29.58$	$t = 26.0$
after "	= 48.2529	$h = 29.76$	$t = 23.5$
Gain in weight	= 0.0030	+ 0.18	— 2.5

Weight of H_2SO_4 tube,

before combustion	= 62.3959	$h = 29.58$	t 26.5
after "	= 66.1076	$h = 29.75$	$t = 23.5$
Gain in weight	= 3.7117	+ 0.17	− 3.0

The correction to vacuum for 3.7117 grams of water weighed with brass and platinum weights is 4.1 mg.

Gain in weight of H_2SO_4	= 3.7117	grams.
" " P_2O_5	= 0.0030	"
Correction to vacuum	= 0.0041	"
" for t and h, P_2O_5	= 0.0004	"
" " " H_2SO_4	= 0.0005	"
Total H_2O formed	= 3.7197	"
Weight H taken	= 0.4144	"
Weight O combined	= 3.3053	"

$$\text{Atomic weight of Oxygen} = \frac{2 \times 3.3053}{0.4144} = 15.953$$

% H in water = 11.140. % O in water = 88.860.

SERIES III. *Determination* 5.

Weighings of the globe: —

					Grams.
Exhausted.	Nov.	8th,	7.45 A. M.	Tare	= 0.1127
	"	"	11.10 A. M.	"	= 0.1125
	"	"	12.00 M.	"	= 0.1122
	"	"	5.00 P. M.	"	= 0.1122
	"	9th,	8.40 A. M.	"	= 0.1121
	"	"	9.15 A. M.	"	= 0.1119
	"	"	12.40 P. M.	"	= 0.1120
	"	"	4.00 P. M.	"	= 0.1121
	"	10th,	8.40 A. M.	"	= 0.1119
	"	"	10.40 A. M.	"	= 0.1120
	"	11th,	8.40 A. M.	"	= 0.1121
	"	"	10.15 A. M.	"	= 0.1120
			Average	"	= 0.1120

			Grams.
Filled with Hydrogen.	Nov. 11th, 12.45 P. M.	Tare	= 0.5325
	" " 4.00 P. M.	"	= 0.5348
	" " 6.00 P. M.	"	= 0.5328
	" 12th, 12.00 M.	"	= 0.5273
	" 14th, 8.00 A. M.	"	= 0.5273
	" " 9.30 A. M.	"	= 0.5273
	" " 11.15 A. M.	"	= 0.5273

Tare filled with H = 0.5273
" empty = 0.1120
Weight H = 0.4153

The combustion was started at 11.25 A. M., and stopped at 6 P. M.

Weight of P_2O_5 tube,

before combustion	= 48.1795	$h = 30.03$	$t = 16.5$
after "	= 48.1832	$h = 30.00$	$t = 18.0$
Gain in weight	= 0.0037	-0.03	$+1.5$

Weight of H_2SO_4 tube,

before combustion	= 64.9625	$h = 30.03$	$t = 16.5$
after "	= 67.6832	$h = 30.00$	$t = 18.0$
Gain in weight	= 3.7207	-0.03	$+1.5$

Gain in weight of H_2SO_4 = 3.7207 grams.
" " P_2O_5 = 0.0037 "
Correction to vacuum = 0.0041 "
3.7285 "

Correction for t and h, H_2SO_4 = −0.0002 "
" " " P_2O_5 = −0.0002 "

Total H_2O formed = 3.7281 "
Weight H taken = 0.4153 "

Weight O combined = 3.3128 "

$$\text{Atomic weight of Oxygen} = \frac{2 \times 3.3128}{0.4153} = 15.954$$

% H = 11.139. % O = 88.861.

ATOMIC WEIGHT OF OXYGEN.

Table of Final Results.

SERIES I.

Weight of Hydrogen.	Weight of Water.	Atomic Weight of Oxygen.
0.4233	3.8048	15.977
0.4136	3.7094	15.937
0.4213	3.7834	15.960
0.4163	3.7345	15.941
0.4131	3.7085	15.954

Average = 15.954 ± 0.0048

SERIES II.

0.4112	3.6930	15.962
0.4089	3.6709	15.955
0.4261	3.8253	15.955
0.4197	3.7651	15.942
0.4144	3.7197	15.953

Average = 15.953 ± 0.0022

SERIES III.

0.42205	3.7865	15.943
0.4284	3.8436	15.944
0.4205	3.7776	15.967
0.43205	3.8748	15.937
0,4153	3.7281	15.954
0.4167	3.7435	15.967

Average = 15.952 ± 0.0035

Total average = 15.953 ± 0.0017

Dumas's value = 15.960 ± 0.0070

On examining the table, it will be noticed that the mean of the determination by the electrolytic method is the mean of all the determinations combined, and that the probable error of the total average is only about one fourth as great as the error of the nineteen determinations of Dumas, which are incomparably the best that have hitherto been made.

It does not now seem possible to escape from the conclusion, that the proportions in which the purest hydrogen that can be made combines with oxygen to form water are those of 2 to 15.953, with a possible error far within the $\frac{1}{100}$ of a single unit.

The question, of course, still remains, Is the hydrogen thus prepared the typical hydrogen element? But this is the same question which must arise in regard to any one of the elementary substances; and all that we can say is, that the evidence in regard to the purity of the hydrogen we have used is as good as any that can be adduced in regard to any one of the elementary substances whose atomic weight has been most accurately determined. The question as regards Prout's hypothesis narrows itself now to this one point; and here we must be content to leave it until further investigation has given us more knowledge in regard to the nature of elementary substances.

The writer at first planned to carry out the investigation on a much larger scale, and for the purpose had blown a globe similar to that represented by Fig. 1, but of five times the capacity, and counterpoised it by the same general method. This globe held twenty-five litres (somewhat over two grams of hydrogen gas), or five times as much as the globe actually used; but the difficulties of carrying out the determinations on this scale led him to reduce the scale of the determinations to that actually adopted; and in the view of the results finally reached, it is evident that no appreciable advantage would have been gained from the enormous expenditure of time and labor which the process on a large scale involves. Assuming that the difficulties of preparing pure hydrogen gas on that scale could have been overcome, it would have required from five to seven hours to fill the globe, and four or five days continuously to complete the combustion.

Moreover, after many trials, the writer could not procure a globe that would stand the requisite pressure weighing less than two and one half kilograms, and with this weight and volume it was not possible, with the best balance he could command, to distinguish half a milligram with as much accuracy as he could one tenth of a milligram with the smaller apparatus, while a vastly longer time was required to reach equilibrium. A great deal of time was spent in endeavoring to perfect this larger apparatus, and a very thorough knowledge was acquired of its relative efficiency. The greatest gain that could have been expected in carrying out the work on this scale would have been the reduction of the probable error to about one half of the present amount, but it is obvious that this gain could be of no importance in the present condition of the science. The accuracy we have reached is far beyond the demands of any analytical work; and, as we have shown, the theoretical question in regard to Prout's law has been settled so far as analytical work can solve the problem. It now turns solely on the typical character of the material we call hydrogen, when prepared in the purest condition known to modern science.

In considering the bearing of the result now published on Prout's hypothesis, it must be borne in mind that it confirms in a most striking manner the result of Dumas, based on the weight of oxygen which water contains, and in connection with his results furnishes a complete analysis of water, with a degree of accuracy as great as can be expected, or as has ever been obtained, in any analytical work.

Complete Analysis of Water.

Percentage of Oxygen after Dumas	88.864 ± 0.0044
Percentage of Hydrogen according to the present work	11.140 ± 0.0011
	100.004 ± 0.0045

It must be remembered that in Dumas's investigation the oxygen alone was weighed, while in the present investigation the hydrogen alone was weighed, and the fact that these two wholly independent analytical results made under such widely different circumstances exactly supplement each other within the limits of probable error, is an evidence of accuracy and a proof of finality which is irresistible.

It would have been highly desirable, if it had been possible, to determine both the oxygen and the hydrogen in one and the same analytical process, as the writer succeeded in doing in the case of silver, bromine, and antimony, and he made many experiments on the reduction of oxide of silver by hydrogen with this view. He succeeded in preparing pure oxide of silver, of definite composition, but the investigation was interrupted by the failure of his sight before he was able to overcome the grave experimental difficulties which the process presented. In view, however, of the present results, it is doubtful whether any advantage would have been gained by that mode of experimenting, for no more certain confirmation could have been reached than that furnished by a comparison of Dumas's results with those of this paper.

Since this investigation was essentially finished, and the results communicated to the American Academy at their meeting of June 15, 1887, we have received from the author a "Sonderabdruck" from the "Berichte der Deutschen Chemischen Gesellschaft," dated the 26th of July following, and entitled: "E. H. Keiser: Ueber die Verbrennung abgewogener Mengen von Wasserstoff und über das Atomgewicht des Sauerstoffs." In this paper Mr. Keiser distinctly recognizes the importance of directly weighing the hydrogen in the determination of the atomic weight of oxygen, and quotes the remarks of Dumas given above. He has also devised a very ingenious method of weighing hydrogen when occluded by palladium; but the preliminary results he publishes are far from having the degree of accuracy required, and lead us to infer that, like our own preliminary results, they must be vitiated by varying impurities in the hydrogen gas used. The three determinations whose results he publishes gave for the atomic weight of oxygen respectively 15.873, 15.897, and 15.826.

We are sorry if Mr. Keiser has entered on somewhat the same field which we have so long occupied without knowledge of our work. But, as above stated, our investigation was begun more than five years ago; and the methods employed have been freely explained to the many chemists, both American and European, who have visited Cambridge during the interval. We earnestly hope that Mr. Keiser will carry out his investigation; for so important a constant as the atomic weight of oxygen cannot be too often verified.

J. P. C. CAMBRIDGE, December 15, 1887.

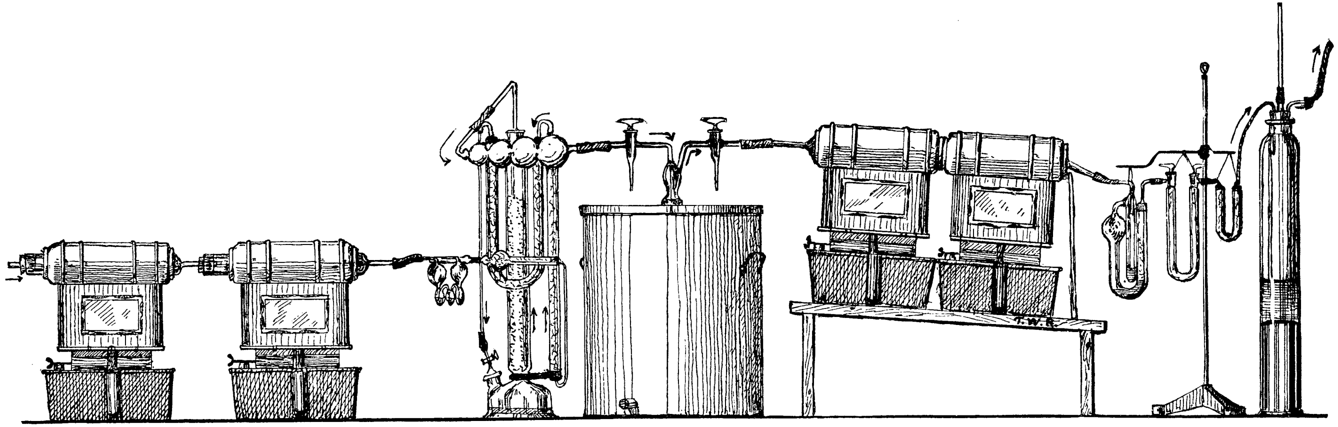

Fig. 6.

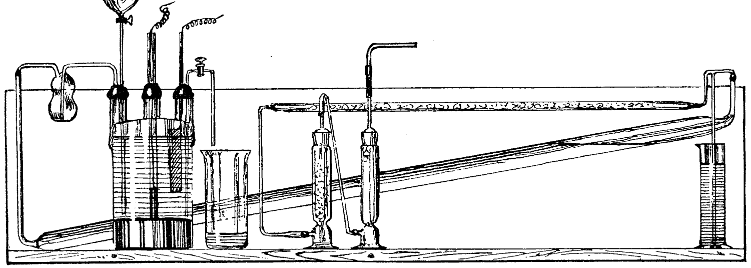

Fig. 7.

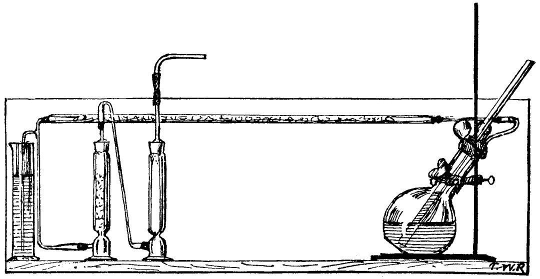

Fig. 8.